

Stochastic Thermodynamics for Inhomogeneous Media

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A unifying framework for the thermodynamics of fluctuating systems with Fokker-Planck dynamics has been developed by Seifert using the notion of stochastic entropy. Here we consider the extension of this formalism to the case of *inhomogeneous media*, where the diffusivity $D = D(x)$ is state-dependent (multiplicative noise) and the usual fluctuation theorems can be violated. We introduce to this effect the concept of *relative stochastic entropy*, and use it to generalize (i) the maximum-entropy principle for the Gibbs canonical ensemble, (ii) the second law of thermodynamics and (iii) Seifert’s integral fluctuation theorems. Our “relative stochastic thermodynamics” can be used e.g. to describe the stochastic motion of colloidal particles dragged in viscous fluids with space-dependent viscosity and/or temperature.

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Introduction. While classical thermodynamics was originally designed for macroscopic systems described by a handful of state variables, it is now well understood that concepts such as energy and entropy are also relevant to mesoscopic non-equilibrium processes where thermal fluctuations play an essential rôle [1, 2]. Following this line of thought, a number of fluctuation theorems (FT) have been derived in the last decade [3–5] (see [2] for a up-to-date review), and their relevance to experimental setups such as the dragging of colloidal particles in viscous fluids [6] demonstrated by several authors [7–9]. The key elements of these theorems are (i) a bath at thermal equilibrium, (ii) a mesoscopic system with (deterministic or stochastic) dynamics and (iii) a suitably defined, protocol-dependent notion of entropy for the system and its surrounding medium. Generally speaking, one then shows that [2]

$$\langle e^{-\Delta s} \rangle = 1, \quad (1)$$

where Δs is the total entropy variation along the protocol, and the brackets denote an average over non-equilibrium fluctuations. This identity implies in particular $\langle \Delta s \rangle \geq 0$, which is the mesoscopic version of the second law of (non-equilibrium) thermodynamics.

The integral FT (1) has been obtained by Seifert in the context of Fokker-Planck dynamics [10]. Elaborating on the work of Sekimoto [11], Crooks [5] and Qian [12], he considers the overdamped motion of a particle in a thermal bath (a viscous fluid), and defines a trajectory-dependent entropy as the sum of two terms: the “system entropy” s_s measuring the uncertainty on the instantaneous position of the particle, and the “medium entropy” s_m associated to the heat dissipated in the medium as an

external driving force is applied to the particle. In this setup, the total entropy $s = s_s + s_m$ can be showed to satisfy the relation (1), where the average is over Brownian trajectories, for *arbitrary* initial conditions and time-dependent driving.

One key assumption underlying this result is that the medium be *homogeneous*, so that the diffusivity D is constant. The purpose of this paper is to address the fate of stochastic thermodynamics when this assumption is dropped. Inspired by our recent discussion of the “geometric ratchet effect” [13], where the FT (1) is violated because the *free* equilibrium particle distribution p_\emptyset^* (that is, without external driving) is *inhomogeneous*, we argue that the suitable notion of entropy for inhomogeneous media is not the standard Gibbs form $-\int dV p \ln p$, but rather the *relative entropy* of p with respect to p_\emptyset^* . With this key concept on hand, we can generalize both the canonical equilibrium distribution and the Seifert integral FT for non-equilibrium processes to inhomogeneous media.

The use of relative entropy is certainly not new to this work. Originally introduced in the context of mathematical statistics [14], relative entropy has been used as a useful tool in many contexts, from quantum information [15] to black hole physics [16]. What we show here is that, in inhomogeneous media, relative entropy is not merely a useful mathematical tool; it is *the* thermodynamic entropy driving irreversible processes. The author did not find this claim spelled out explicitly in the literature, although hints to this effect can be found in the early works [17] and [18], and more recently in [19, 20]. In particular, our use of the concept of relative entropy is not to be confused with the “phase-space perspective” of Kawai,

Parrondo and Van den Broeck [21], as will be explained in the conclusion.

Inhomogeneous Fokker-Planck dynamics. Consider an overdamped particle with mobility $\mu(x)$ diffusing in an inhomogeneous thermal bath, subject to a protocol-dependent force [10]

$$F(x, \lambda) = -\nabla V(x, \lambda) + f(x, \lambda). \quad (2)$$

Here the “protocol” $\lambda(t)$ is an external control parameter varied continuously on the time interval $t_i \leq t \leq t_f$, $V(x, \lambda)$ is a local conservative potential and $f(x, \lambda)$ is a force applied directly to the particle. The trajectory $x(t)$ of the particle is governed by the Langevin equation

$$\dot{x} = \mu(x)F(x, \lambda) + \zeta(x), \quad (3)$$

where $\zeta(x)$ is a normalized Gaussian white noise with local covariance $\langle \zeta^i(x, t) \zeta^j(x, t') \rangle = 2D(x) \delta_{ij} \delta(t - t')$. We assume that the local diffusivity $D(x)$ is related to the local mobility $\mu(x)$ and temperature $T(x)$ according to Einstein’s relation $D(x) = \mu(x)T(x)$. We leave the dimension of the configuration space d unspecified.

As is well known [22], the Langevin equation (3) is not well defined as such because of a discretization ambiguity in the definition of the velocity \dot{x} . Depending on the actual nature of the medium’s inhomogeneity, this ambiguity results in one of several inequivalent Fokker-Planck equations for the probability density p . In this paper we focus on those systems where the microscopic physics dictates to interpret (3) in the Itô sense,

$$dx = \mu(x)F(x, \lambda)dt + \sqrt{2D(x)}dW, \quad (4)$$

or equivalently by

$$\circ dx = (\mu(x)F(x, \lambda) - \nabla D)dt + \sqrt{2D(x)} \circ dW \quad (5)$$

where $\circ d$ is the Stratonovich stochastic differential and W the standard d -dimensional Wiener process. Both equations (4) and (5) correspond to the Fokker-Planck equation

$$\partial_t p + \nabla \cdot j = 0 \quad (6)$$

with the probability current

$$j \equiv \mu F p - \nabla(Dp). \quad (7)$$

Our motivation for considering this case in detail¹ is because *its violates the Seifert FT*—and indeed any statement to the effect that “the entropy of an isolated system at local equilibrium can never decrease”. This is

¹ Rather than for instance $j = \mu F p - D \nabla p$, as in [24].

due to the following fact: the equilibrium distribution for (6) is inhomogeneous *also in the absence of any external forcing* ($F = 0$). Instead, the free equilibrium distribution $p_{\mathcal{O}}^*$ satisfies $p_{\mathcal{O}}^*(x) \propto D(x)^{-1}$. This implies for instance that, when $D(x)$ has a trap-like shape (viz. grows monotonously about a global minimum), stochastic dynamics can result in “anti-diffusion”: initially spread Brownian particles will spontaneously concentrate at the bottom of the trap *without any external force*, hence $\langle \Delta s \rangle < 0$; see Fig. 1 for an explicit example of this effect.

Let us emphasize that systems where transport is well described by equations (4) and (6), and in particular where the free equilibria $p_{\mathcal{O}}^*$ are not constant, are not mathematical abstractions: they do exist in nature. Among such systems, we can mention fusion plasmas [25], thermodiffusion at certain values of the Soret coefficient [26], or more simply fluid mixtures with variable viscosity [23]. Considering the Einstein relation $D = \mu T$, we see that the presence of temperature gradients, as in thermodiffusion, is not the only source of inhomogeneity: a *mobility gradient* at fixed temperature will result in $\nabla D \neq 0$ as well. The fluid system of [23] is an instance of this second possibility. This case is particularly interesting, for it shows that with a viscosity profile decreasing monotonously about a maximum, Brownian particles can *self-organize* (viz. decrease their mean entropy) without any heat being exchanged with the environment; see [13] for an in-depth discussion of this surprising phenomenon.²

Relative stochastic entropy. What this “geometric ratchet effect”—as we proposed to call it in [13]—shows is that the ordinary Gibbs entropy of the particle distribution is not the relevant quantity to describe dissipative processes in inhomogeneous media. As argued in [13] (and already understood in [17, 18]), one should use instead the *relative entropy* of p with respect to the free

² Another case where (6) with (7) naturally arises is in general relativity. Indeed, the diffusion equation for a Brownian particle in local equilibrium with a fluid in hydrostatic equilibrium in a gravitational field is [27]

$$\partial_t = D_0 \Delta(Np), \quad (8)$$

where N is the lapse function in the rest frame of the fluid and D_0 is the proper (constant) diffusivity. This is of course the Fokker-Planck equation (6) with $F = 0$. To our knowledge, this connection between inhomogeneous diffusion and general relativity is the first realization of the “analogue gravity” paradigm [28] in the realm of dissipative physics.

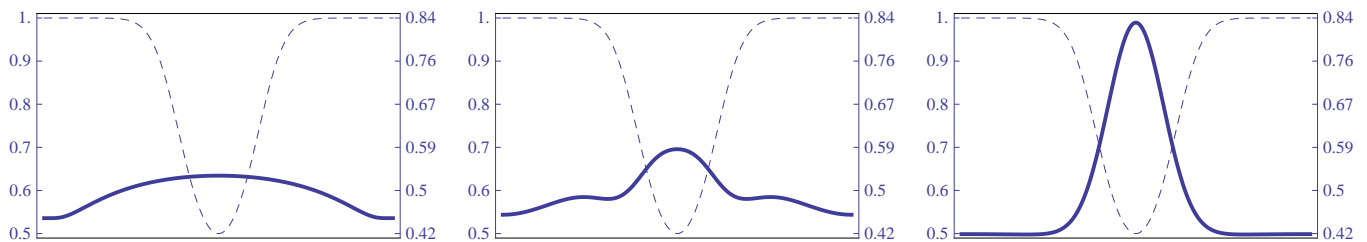


FIG. 1: Numerical solution of the one-dimensional FP equation (6) with $F = 0$ and Neumann boundary conditions, at three times $t_0 < t_1 < t_2$ (left to right). The dashed curve (left axis) is the local diffusivity $D(x)$ normalized to its maximal value; the thick curve (right axis) are the probability densities $p(t_i, x)$, with x in arbitrary units. The amplitude of variation of $D(x)$ is consistent with the fluid experiment of [23]. The conventional Gibbs entropy manifestly decreases in this setup, see also [13].

($F = 0$) equilibrium $p_{\mathcal{O}}^*$, namely

$$K(p \| p_{\mathcal{O}}^*) \equiv - \int p \ln \frac{p}{p_{\mathcal{O}}^*} dv, \quad (9)$$

where dv is the volume element. This quantity, also known as the Kullback-Leibler divergence [14], measures the informational “distance” between the instantaneous distribution p and the equilibrium $p_{\mathcal{O}}^*$.

That $K(p \| p_{\mathcal{O}}^*)$ is a suitable measure of irreversibility can be seen by checking that, unlike the standard Gibbs-like entropy $-\int dV p \ln p$, the relative entropy $K(p \| p_{\mathcal{O}}^*)$ can *never decrease*³ in a free diffusion process: starting from the driftless Fokker-Planck equation $\partial_t p = \Delta(Dp)$, one checks easily that [17, 18]

$$\frac{dK(p \| p_{\mathcal{O}}^*)}{dt} = \int \frac{(\nabla(Dp))^2}{Dp} dv \geq 0. \quad (10)$$

Of course, the standard Gibbs entropy is a particular case of relative entropy, namely the relative entropy with respect to a *uniform* prior.

Relative canonical ensemble. The first implication of the fact the free equilibrium distribution $p_{\mathcal{O}}^*$ is inhomogeneous is the modification of the canonical ensemble for thermal equilibrium ($f = 0$, $T = \text{const}$). Indeed, adapting Jaynes’ informational reasoning [29] to this setup, one is led to the *principle of maximum relative entropy*: the

equilibrium canonical distribution p^* in a given potential is the one maximizing the entropy relative to the free equilibrium $p_{\mathcal{O}}^*$, subject to the mean energy constraint $\int dx p^* V = \text{const}$. Denoting as usual $\beta = 1/T$ the corresponding Lagrange multiplier leads to the variational problem

$$\max \left(- \int p \ln \frac{p}{p_{\mathcal{O}}^*} dv + \beta \int p V dv \right), \quad (11)$$

which is equivalent to

$$\max \left(- \int p \ln p dv + \beta \int p \tilde{V} dv \right), \quad (12)$$

where $\tilde{V} = V - \ln p_{\mathcal{O}}^*/\beta$ plays the rôle of a *generalized potential* [18].⁴ The solution of this variational problem defines the *relative canonical ensemble*,

$$p^* \propto e^{-\beta \tilde{V}} = p_{\mathcal{O}}^* e^{-\beta V}, \quad (13)$$

which is the suitable one for thermal equilibrium in inhomogeneous media.

Second law of thermodynamics. Let us now come back to the problem of driven diffusion. As stressed by Seifert [10], the entropy variation of stochastic systems splits in two terms: the variation of the system entropy Δs_s , associated to the uncertainty on the instantaneous position $x(t)$ of the system, and the entropy dissipated in the medium Δs_m , associated to heat transfers between the work received by the system and the surrounding medium. In the context of Fokker-Planck dynamics, these

³ Incidentally, the first use of relative entropy to resolve an apparent violation of the second law of thermodynamics was perhaps by Gibbs himself: adding a term $\ln N!$ to the entropy of a gas of indiscernible particles is really considering the entropy relative to the equilibrium distribution $p_{\mathcal{O}}^* \propto 1/N!$ over the redundant N -particle phase space Γ_1^N (here Γ_1 is the 1-particle phase space).

⁴ The gradient $\nabla \ln p_{\mathcal{O}}^*/\beta$ is of course the same as the Stratonovich correction in equation (5).

quantities can be defined for a single trajectory $x(t)$ [10]. Following our discussion of relative entropy, we define the *relative system entropy* by

$$s'_s(t) \equiv -\ln \frac{p(x(t), t)}{p_{\mathcal{O}}^*(x(t))}. \quad (14)$$

We define furthermore the *dissipated entropy* in the medium by the Stratonovich integral

$$\Delta s_m \equiv \int_{t_0}^{t_1} \frac{F(x(s), \lambda(s))}{T(x(s))} \cdot \circ dx(s). \quad (15)$$

The *total relative stochastic entropy* is then the sum

$$s' \equiv s'_s + s_m. \quad (16)$$

Its variation $\circ ds'(t)$ along the stochastic trajectory $x(t)$ is given by

$$\circ ds'(t) = -\frac{\partial_t p}{p} dt - \frac{\nabla(p/p_{\mathcal{O}}^*)}{p/p_{\mathcal{O}}^*} \cdot \circ dx(t) + \frac{F}{T} \cdot \circ dx(t), \quad (17)$$

with each term is evaluated at $x(t)$. Using (7) and the Einstein relation, this gives

$$\circ ds' = -\frac{\partial_t p}{p} dt + \frac{j}{Dp} \cdot \circ dx, \quad (18)$$

which is the generalization of Seifert's "first central result" in [10] to inhomogeneous media. It provides an explicit formula for the entropy production along a single stochastic trajectory. Paraphrasing [10], we can then use (18) to obtain the *mean* total entropy production rate

$$\left\langle \frac{ds'}{dt} \right\rangle = \frac{dK(p \| p_{\mathcal{O}}^*)}{dt} \geq 0. \quad (19)$$

This inequality is the proper statement of the second law of non-equilibrium thermodynamics in the inhomogeneous context. Again, its validity relies crucially on the use of the *relative* entropy of the particle with respect to their free equilibrium distribution, and *not* the standard Gibbs expression.

Relative fluctuation theorem. We now move on to consider the relative version of Seifert's FT. Following the standard procedure [2], we proceed by computing the ratio of the probability of a given trajectory $x(t)$ starting at x_0 and ending at x_1 to that of the reversed trajectory $\tilde{x}(t) = x(\tau - t)$ driven by the reversed protocol $\tilde{\lambda}(t) = \lambda(\tau - t)$; the integral FT then follows from a suitable weighting of the initial position and final position of the path.

In the standard setup, the probabilities $P[x(t)]$ and $P[\tilde{x}(t)]$ of a path $x(t)$ and its reverse $\tilde{x}(t)$ satisfy

$$\ln \frac{P[x(t)]}{P[\tilde{x}(t)]} = \Delta s_m, \quad (20)$$

i.e. the irreversibility of a path is measured by the entropy dissipated in the medium; in the limit where $F = 0$, this gives $P[x(t)] = P[\tilde{x}(t)]$. In the inhomogeneous media considered in this paper, this it is clear that this relation cannot hold anymore: a path ending at point where the free equilibrium probability $p_{\mathcal{O}}^*$ is larger must be favored over the reversed path.

Indeed, using the generalized Onsager-Machlup function [30, 31]

$$L(x, \dot{x}, \lambda) = \frac{(\dot{x} - \mu(x)F(x, \lambda) - h(x))^2}{4D(x)} + f(x) \quad (21)$$

where $h(x) = (d/2 - 1)\nabla D(x)$ and $f(x)$ is a function of x only which we do not need to specify, we can write the path probability density $\pi[x(t)]$ with respect to the generalized Wiener measure [22]

$$\mathcal{D}[x(t)] = \lim_{N \rightarrow \infty} \prod_{n=0}^{N-1} \left(4\pi\tau D(x_n) \right)^{-d/2} dv(x_n) \quad (22)$$

as

$$\pi[x(t)] = \exp \left(- \int_{t_0}^{t_1} L(x(t), \dot{x}(t)) dt \right) \delta(x(0) - x_0). \quad (23)$$

Here $\tau = (t_1 - t_0)/N$, $x_n = x(t_0 + n\tau)$ and \dot{x} is the time-derivative of a smooth trajectory approximating $x(t)$. Hence

$$\ln \frac{P[x(t)]}{P[\tilde{x}(t)]} = -\frac{d}{2} \ln \frac{D(x_1)}{D(x_0)} - \int_{t_0}^{t_1} \left(L(x(t), \dot{x}(t), \lambda(t)) - L(\tilde{x}(t), \dot{\tilde{x}}(t), \tilde{\lambda}(t)) \right) dt \quad (24)$$

Evaluating the integral explicitly and using again $p_{\mathcal{O}}^* \propto D^{-1}$, we arrive at

$$\ln \frac{P[x(t)]}{P[\tilde{x}(t)]} = \Delta s_m(\tau) + \ln \frac{p_{\mathcal{O}}^*(x_1)}{p_{\mathcal{O}}^*(x_0)}. \quad (25)$$

Note the extra term with respect to the homogeneous case [10].

To derive the corresponding integral FT, we weigh the starting point x_0 by the initial condition $p(x_0, t_0)$ and the

final point $\tilde{x}_0 = x_1$ by the evolved distribution $p(x_1, t_1)$, and form the ratio

$$R = \ln \frac{P[x(t)] p(x_0, t_0)}{P[\tilde{x}(t)] p(x_1, t_1)}. \quad (26)$$

According to (25), R is nothing but the variation of the total *relative* stochastic entropy, viz. $R = \Delta s'$. We can then apply the classic identities

$$\langle e^{-R} \rangle = \int \mathcal{D}[x(t)] \pi[x(t)] p_0(x_0, t_0) e^{-R} \quad (27)$$

$$= \int \mathcal{D}[\tilde{x}(t)] \pi[\tilde{x}(t)] p(\tilde{x}_0, t_1) = 1, \quad (28)$$

to obtain

$$\langle e^{-\Delta s'} \rangle = 1. \quad (29)$$

This relation is the main result of this work. Note that choosing a more general weight $p_1(x_1)$ for the final point x_1 (indeed any probability density) would have led instead to the generalization of Seifert's "unifying" FT [10]

$$\left\langle e^{-\Delta s_m} \frac{p_1(x_1)}{p_\emptyset^*(x_1)} \frac{p_\emptyset^*(x_0)}{p(x_0, t_0)} \right\rangle = 1. \quad (30)$$

Conclusion. We have extended the framework of stochastic thermodynamics to state-dependent diffusions, with an emphasis on the notion of relative stochastic entropy. We have showed that the latter is the general measure of irreversibility for inhomogeneous media, and not the ordinary Gibbs entropy.

It is important to contrast our use of this concept from the one made in [21]: while Kawai *et al.* consider the relative entropy $K(\rho \parallel \tilde{\rho})$ of the probabilities ρ and $\tilde{\rho}$ for the forward and backward processes, our relative stochastic entropy s'_s involves the forward process and the free equilibrium state. This is conceptually very different; in particular, the framework of [21] does not apply to inhomogeneous media. A much closer application of this concept is in [20] and [19], where the entropy of a state relative to the corresponding equilibrium state is discussed; the main difference with the present work is that, in both papers, the gradients of the equilibrium distributions are always due to *energy* variations. Here we also considered ∇D as a source of inhomogeneity of p^* .

Let us emphasize that although we focused on the Itô interpretation (4) of the Langevin equation, our analysis is not restricted to this case: in a setup where (3) has a homogeneous free equilibrium, our results reduce to the standard ones, with relative entropy coinciding with ordinary entropy [24]. We also note that our framework

can be immediately adapted to Markovian dynamics on a discrete state space, as in [10].

To put our extended framework to test, one could for instance repeat the famous colloidal particle dragging experiment [7], but now within a fluid with varying viscosity, as in [23].

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